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Comparison of HEPA Filter Test Methods
in Corrosive Environments

MASTER

L. P. Murphy, S. J. Fernandez and B. G. Motes

Exxon Nuclear Idaho Company
Idaho Chemical Programs - Operations Office
Idaho National Engineering Laboratory

ABSTRACT

The in-plant testing of process off-gas HEPA filters is an important quality control activity of the ALARA (As Low As Reasonably Achievable) policy at nuclear facilities. Imprecise and irreproducible data were recorded during DOP testing at the Atmospheric Protection System (APS) of the Idaho Chemical Processing Plant (ICPP). The tests at the APS are performed in an environmental that has high humidity, high temperatures and has NO_x present.

An evaluation of three HEPA filter test methods in corrosive environments was conducted: the dioctyl phthalate (DOP) method (US Standard Method ANSI N-101.1-1979) the sodium chloride method (British Standard 3928:1969), and the soda-fluorescein or uranine method (French Standard AFNOR STD NFX 44-011). The effects of high humidity, temperature and oxides of nitrogen (NO_x) on each method was examined. The effects of each variable and any interaction between variables on the test methods were examined. Recommendations for changes in the standard methods to reduce erratic on-line results are presented.

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I. INTRODUCTION

The Idaho Chemical Processing Plant (ICPP), located at the Idaho National Engineering Laboratory, is operated by Exxon Nuclear Idaho Company for the Department of Energy to recover uranium from spent fuel elements. Aqueous wastes from the recovery process are solidified in a fluidized bed waste calcining facility.

The Waste Calcining Facility (WCF) produces an off-gas stream that contains high concentrations of nitrogen oxides (NO_x), and has high temperatures and high relative humidities. The final process off-gas clean up system before emission to the environment is the Atmospheric Protection System (APS). The APS consists of a condensor, demistor, superheater, prefilter, and finally three HEPA banks. An isometric view of the APS process off-gas area is shown in Figure 1.

To ensure that the HEPA filters are functioning properly, it is necessary to ascertain whether these filters have sustained damage during transportation to or installation in the APS and that the assembly of the filter elements is satisfactory. Thus, testing these HEPA filter banks in place in the corrosive environment is required.

To provide the most appropriate filter test method, the American DOP, the British NaCl and the French uranine filter test methods were evaluated in the laboratory to determine the combined effects of temperature, humidity, and NO_x . The most appropriate method as determined by this laboratory evaluation was demonstrated in-plant.

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Figure 1. Isometric View of APS Process Off-Gas Area

Insert Figure 1

II. DESCRIPTION OF THE TEST METHODS

The development of HEPA filter test methods has been conducted in several countries. Some, such as the American DOP method use liquid droplets as a challenge aerosol, while others such as the British NaCl and French uranine methods use solid particles. The Nuclear Energy Agency of the Organization for Economic Cooperation and Development, recommended ⁽¹⁾ at a meeting for the exchange of information on gaseous wastes, that test conditions for HEPA filters be defined with respect to high temperatures, humidity and radiation conditions. The participants at this meeting considered the DOP, uranine, and sodium chloride methods to be most important and recommended that these methods be investigated as to suitability for in-situ tests in all process stream conditions. Brief descriptions of these methods are given in the following sections. More detailed descriptions are presented elsewhere. ^(2,3, 4)

A. Diethyl Phthalate (DOP)

This method was originally developed in the United States and employs either a monodisperse or polydisperse liquid aerosol of DOP. The aerosol can be produced by either a thermal or pneumatic type generators.

An aerosol at a concentration of 100 mg/M^3 from either type of generator is introduced upstream of the filter to be tested. To determine the filter efficiency, the air is sampled before and after the filter and the efficiency is determined by comparing particle concentrations upstream and downstream of the filter. These particle concentrations are measured with a forward light scattering photometer. The DOP method is useful when there are many tests to be performed and/or no interferences are present since the penetration measurements are practically instantaneous.

B. Sodium Chloride (Sodium Flame)

In the sodium flame test, performed according to British Standard 3928, an aqueous solution of sodium chloride is atomized. The moisture evaporates, leaving a dry cloud of solid cubic salt particles. These particles range from 0.01 to 1.7

μm , with a mass median diameter of $0.6 \mu\text{m}$. To measure filter efficiency, samples of the air before and after the filter are directed to a flame photometer which measures the intensity of the characteristic yellow sodium emission line produced in a hydrogen flame. The intensity of this emission line is directly proportional to the mass of sodium chloride present.

This method, like the DOP method, measures penetration in real-time.

For our studies, the single particle laser aerosol spectrometer⁽⁵⁾ was substituted for the flame photometer, because the sampling conditions at the APS were not practical for the flame photometer.

C. Soda-Fluorescein (Uranine)

In France, a fluorescent aerosol is used to evaluate HEPA filters. The aerosol is produced by atomizing an aqueous solution of uranine. After the elimination of large droplets with an inertial separator, the remaining liquid aerosol is evaporated by dilution with dry air. A solid aerosol results with an average particle size corresponding to the size of maximum penetration ($0.08 \mu\text{m}$) for HEPA filters at the face velocity encountered at the APS.⁽⁶⁾

The test aerosol is injected upstream of the filter being tested and samples are collected before and after the HEPA on 47 mm membrane filters. The uranine is extracted from the sample filters with ammonium hydroxide and the extracts measured fluorimetrically (496 nm excitation, 517 nm emission). This fluorimetric analysis requires about 45 minutes.

III. LABORATORY EVALUATION

1. Description of Laboratory Tests

A. Experimental Apparatus

Because of the highly radioactive environment of the WCF and APS off-gas, an experimental system was constructed to simulate the off-gas conditions. The studies were conducted with 20 cm x 20 cm x 15 cm (8 inch x 8 inch x 5 7/8 inch) HEPA filters in tandem. Measurements of aerosol concentrations were taken upstream of both filters, between the filters, and after the final filter.

This test assembly is shown in Figure 2. The DOP, NaCl and uranine methods were adapted to the apparatus with appropriate sampling modifications. The system included an aerosol generator, injection and sampling points for water vapor, NO_x and the test aerosol. Also included was a reaction bomb for partial conversion of NO to NO_2 . This reaction provided a final mixture of 50/50 NO/ NO_2 .

The gas flow through the system was measured by linear mass flow transducers and the flow was maintained at 25 scfm. Humidity and NO_x concentrations were measured with a hygrometer^a and chemiluminescent^b analyzer, respectively. The temperature was monitored with thermocouples and controlled with heat tapes.

B. Experimental Design

The experimental design used in this study was a two-level factorial design containing three variables (temperature, humidity, and NO_x). Factorial designs are, in general composed by selecting a fixed number of levels of each variables, and performing all the possible unique tests. The greatest strength of a factorial design is that the controlled variables can be varied simultaneously, thereby providing information about interaction of variables.

^a Series 1200 AP Optical Dew Point Hygrometer manufactured by General Eastern Corp.

^b Model 10, NO- NO_2 - NO_x Chemiluminescent Analyzer Manufactured by Thermo Electron Corp.

Figure 2 HEPA Filter Test System

Insert Figure 2

The 2^3 factorial design, which is useful for ease of analysis and for its predictive capability for responses that do not have strong curvature in the region of interest, requires that two levels be chosen and all eight ($2^3 = 8$) combinations tested. The two levels chosen for this study were: for temperature, 66 and 93°C; for humidity, 50 and 95%; and for NO_x concentration, 500 and 10,000 ppm.

2. Results and Discussion of Laboratory Evaluation

A. Dioctyl Phthalate (DOP)

The DOP studies were conducted by measuring penetration with the conventional forward light-scattering photometer and the laser single particle aerosol spectrometer. The method used followed as closely as possible that detailed in ANSI N-101.1-1979. When testing streams with high dew points (above 60°C) a drying tube was used to lower the sample dewpoint without removing the aerosol. This dryer is based on selective permeation of water vapor as described by Fernandez, et al.⁽⁷⁾ The effects of humidity, NO_x , and temperature on the standard DOP in-place test method were examined. The results of these examinations were reported at a previous Air Cleaning Conference⁽⁸⁾, however they are discussed briefly in the following sections.

a. Light-Scattering Photometer

Preliminary tests were performed to establish baseline behavior for the photometer by measuring DOP concentrations in room air. The tests were performed at a flow of 25 scfm and temperatures of 66 and 93°C. Each test lasted two minutes and 100 mg/M³ of DOP challenged the filters.

The results of these measurements indicated a background penetration of 0.002% $\pm$.001% (0.2 parts per ten thousand (ppt)).

After the baseline measurements were completed, scoping tests with DOP and conditions ranging from 16% to 50% relative humidity (RH) and from 500 to 10,000 ppm NO_x were performed. The apparent penetrations for all of the tests at 50% relative humidity varied rapidly from 0% to 40% penetration. When measurements were attempted with room air and 500 to 10,000 ppm of NO_x , a negative reading was obtained.

Although the reason for this behavior is far from obvious, an explanation consistent with the data is that at high humidities the water vapor condensed within the scattering chamber causing unrealistically high penetration values. The negative response during the dry NO_x runs was attributed to absorption of the light by the brown-colored NO_2 . During the high humidity, high NO_x tests, the NO_x was scrubbed from the scattering chamber by the condensation, yielding the same response as the high humidity, no NO_x case.

The problem of condensation occurring in the photometer was solved by installing a drying tube to remove water vapor without removing the aerosol. The NO_x interference was eliminated by making the photometer stray light adjustment while sampling the stream containing the NO_x .

Once these modifications were made, experiments were carried out following the experimental design shown below. The eight test design was duplicated, and the average penetration measured was 1.9 ± 0.9 pptt.

Figure 3

2^3 Experimental Design

Relative Humidity %	NO_x (PPM)	Temperature $^{\circ}\text{C}$
50	500	66
95	500	66
50	10,000	66
95	10,000	66
50	500	93
95	500	93
50	10,000	93
95	10,000	93

The design was performed in random order. Each test point was duplicated.

b. Laser Aerosol Spectrometer

Tests similar to the ones performed with the photometer were performed with the laser aerosol spectrometer. These tests established baseline behavior for the laser by measuring DOP concentrations in dry air.

The results of these tests indicated background penetration of $.02 \pm .03$ pptt for the laser spectrometer in a well filtered gas stream.

The baseline measurements were followed with tests designed to ascertain if there were effects due to humidity or NO_x . The results of these measurements indicated that the laser showed no humidity or NO_x effect, yielding the same penetration as the baseline measurements.

The HEPA filter efficiencies in a corrosive environment using the laser were obtained. The average penetration was 1.7 ± 1.6 pptt.

In summary the statistical analysis of the two-level design at the 95% confidence interval gave the following results. Using the photometer, an effect of $+1.5 \pm 0.2$ pptt was observed due to water vapor. All other effects were insignificant at the 95% confidence interval. This water vapor effect may be due to residual condensation in the scattering chamber.

Using the laser spectrometer, a small temperature effect of 0.30 ± 0.09 pptt was observed. The effects of all other variables were insignificant at the 95% confidence interval. The cause of this effect is unknown but may be due to the fact that the vapor pressure of DOP is sufficiently high at temperatures above 80°C to cause a wicking effect through the filter. In any event, this temperature effect is much less than the observed 1.7 pptt variability in the overall average.

By comparison, the laser spectrometer is superior to the photometer since it is three times more sensitive than the photometer and has no water vapor or NO_x interference.

The variability of the DOP method, i.e., 1.5 pptt, is unacceptable. For example, many acceptable filters are in the 99.97 to 99.95% efficiency region, the added 1.5 pptt penetration bias would cause a good filter to fail the DOP test.

B. Sodium Chloride

British Standard 3928: 1969 calls for a solid challenge aerosol to be generated by atomization of an aqueous solution of common salt from large collision - type atomizers. Subsequently, measurements are made upstream and downstream of the filters with a flame photometer. In this study, the standard procedure was modified by substituting the single particle laser aerosol spectrometer for the flame photometer. The flame photometer was thought to be impractical from safety considerations and the laser allows particle size determinations in addition to concentration.

In these experiments, the challenge aerosol was produced by atomizing a 5% solution of NaCl. The particles were polydisperse and 95% were in the range of 0.15 to 1.0 μm . The entire range was from 0.10 to 1.72 μm .

As with the DOP tests, when experiments based on a 2^3 factorial design were performed, the variable that caused the greatest interference was water vapor. No results were obtained at 95% relative humidity and 95°C due to condensation inside the sample line and spectrometer. Heat tracing the sample lines was not possible due to the laser's temperature limitations. Attempts to remove the water vapor with the drying tube mentioned earlier were successful, but the plateout of aerosol inside the dryer was as high as 16%. This plateout was significantly greater than with DOP, probably due to differences in the particle size distributions.

Under normal operating conditions, the relative humidity at the APS rarely falls below 60%. Consequently, any satisfactory system must be suitable for use in humid gas streams. Upon completion of the two-level experimental design, the water vapor effect was determined to be 5.5 pptt (see Table I). Since the maximum penetration allowed is 5.0 pptt, high humidity conditions will cause a "fail" test no matter the actual efficiency of the filter. One possible explanation was suggested by Dorman⁽⁹⁾ who observed that in tests with humidities above 50%, the NaCl aerosol takes on moisture providing a mechanism for particles to pass through the filter.

TABLE I

Measured Filter Efficiencies in Corrosive Environments (NaCl)

<u>Test Conditions</u>			<u>Penetration</u>
<u>Relative Humidity %</u>	<u>NO_x (PPM)</u>	<u>Temperature °C</u>	<u>Parts/10,000 (pptt)</u>
15	500	25	0.6
55	500	25	5.2
15	5,250	25	0.7
55	5,250	25	6.3
15	500	60	0.5
55	500	60	4.4
15	5,250	60	0.6
55	5,250	60	7.5

An analysis of the data listed above based on a two-level design and the 95% confidence interval indicate the standard deviations for the measurements was ± 0.4 pptt. The minimum significant factor effect was ± 0.5 pptt. If the computed factor effect is twice the absolute value of the minimum significant factor then we conclude that there is an effect.

The effects of NO_x and temperature were insignificant within the 95% confidence limits as were the interactions. These findings are consistent with the DOP experiments that showed that the laser was unaffected by NO_x atmospheres. In addition, the NaCl test method has been recommended for temperatures as high as 400°C.⁽¹⁰⁾

In conclusion, the NaCl test method is not recommended for humid atmospheres due to the hygroscopic nature of the test aerosol.

C. Soda-Fluorescein (uranine)

The French soda-fluorescein or uranine method was the final method evaluated. The standard method for the testing of high efficiency filters in France (AFNOR Standard NFX 44011) was studied under high humidities, high NO_x concentrations and high temperatures. In the standard test, aerosol samplings are collected upstream and downstream of the test filter on 47 mm glass fiber filters. The sample flowrate was 2 to 5 m³/hr. The uranine was extracted with 5%

ammonium hydroxide solution. The fluorescence of the solution was then measured at the characteristic uranine wavelengths (496 nm excitation and 517 nm emission). The ratio of the uranine mass concentration downstream of HEPA filter to the quantity of aerosol found upstream is the measured penetration.

To conduct our laboratory evaluation, a uranine generator, filtration pots, and sampling devices were purchased from Societe Setra, Ormesson, France. A Varian Model SF-330 Spectrofluorometer was used to measure fluorescence.

Preliminary tests were conducted at ambient conditions to check the performance of the equipment, the reproducibility of the generator, and the reliability of the sampling and measurement techniques. The amount of aerosol generated was about 18 ± 2.5 mg/hr. Once the initial adjustments were made the generator functioned without attention for three hours with the only requirement being a 100 psi air supply.

The particle size of the aerosol was measured with a diffusion battery.⁽¹¹⁾ All particles were smaller than $0.2 \mu\text{m}$ with a geometric mean diameter of $0.08 \mu\text{m}$.

Penetration measurements were performed on the same test apparatus as the DOP and NaCl tests.

The average penetration of the uranine method was 3.4 ± 1.0 pptt with a range of 2-5 pptt.

The results of the measurements using a 2^3 factorial design indicated that there was no main effect for relative humidities up to 80%, temperatures up to 90°C and NO_x concentrations up to 10,000 ppm. Also, no detectable interaction between temperature, relative humidity and NO_x was observed. However, when condensation was detected on the sample filter the uranium was leached from the filter. This was eliminated by heating the sample filter significantly above the sample dew point.

This laboratory evaluation revealed the French uranine method was satisfactory for use in humid, high temperature, corrosive atmospheres and that it had the following advantages over the DOP and NaCl filter test methods:

- 1) The uranine aerosol maintains its integrity in temperatures up to 120°C and relative humidities as high as 80%.
- 2) The uranine aerosol is a solid with an average particle size of 0.08 µm. This makes the uranine more representative of the particles that penetrate APS HEPA filters.
- 3) The fluorimetric assay eliminates the interferences from in-leakage of non-fluorescent ambient aerosols.

IV. IN-PLANT DEMONSTRATION

The French uranine method was selected for in-plant demonstration tests. This demonstration was performed in conjunction with another task for extending the uranine method to relative humidities greater than 80%. The uranine and DOP methods were used simultaneously to determine the efficiencies of the HEPA filters at the APS.

The APS process off-gas system consists of a condenser, demister, superheater, glass fiber prefilter bank, and three HEPA filters (see Figure 4). The total flow of the combined process off-gases is limited to 4500 cfm and this flow is split equally between the three HEPAs.

A comparison of DOP and uranine was made under WCF conditions. Table II shows the result of this comparison.

TABLE II
In-Plant Measurement W/DOP and Uranine

Filter #	NO _x Present	Measured Efficiencies	
		DOP	Uranine
OGF-100	YES	—	85%

Measurement with DOP and the photometer were not possible while the calciner was operating. The measurement of that filter was made after the calciner shut down (no NO_x) and it failed the DOP test.

More tests were performed during the WCF shut down. The results are shown in Table III. In these measurements fluorescein and eosin-y were used to attempt to extend the uranine method to relative humidities above 80%.

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Figure 4. APS Process Off-Gas Filters

Insert Figure 4

TABLE III

In-Plant Measurements W/DOP and Other Fluorescent Aerosols*

<u>Filter #</u>	<u>Measured Efficiencies</u>		<u>Eosin-Y</u>
	<u>DOP</u>	<u>Fluorescein</u>	
OGF-100	99.96	99.8 ^b	—
-101	99.94		97.4 ^b
-102	99.98		99.93 ^b

*Note: WCF was shutdown

^a These filters failed the DOP test

^b The difference in the efficiencies between DOP and the fluorescent aerosols is thought to be due to the smaller diameter of these aerosols.

In summary, because imprecise and irreproducible data were recorded during past DOP testing ⁽¹²⁾ at the Atmospheric Protection System during WCF operations, we collected data using the uranine aerosol at this point. But, the WCF has been down for an extended period and the data from in-plant demonstrations has been limited. However, brief operating experience has shown that measurements are possible with uranine at the APS in NO_x containing streams.

V. CONCLUSIONS AND RECOMMENDATIONS

In conclusion, the French uranine method has the following advantages over the NaCl and DOP filter test methods:

- 1) The uranine aerosol is a solid aerosol, rather than a liquid like the DOP aerosol. This makes the uranine aerosol more representative of the solid particles in the WCF off-gas stream.
- 2) The fluorimetry assay used in the uranine method eliminates interferences from in-leakage of non-fluorescent ambient aerosols. The DOP and NaCl methods are subject to these interferences.
- 3) Without modifications the uranine method can be used in relative humidities as high as 80% (with modifications the humidity limits can be extended) and temperatures as high as 120°C. The DOP method is only recommended for ambient conditions; while the NaCl method can withstand temperatures as high as 400°C, it is not recommended for relative humidities above 50%.

The advantages listed above underscore the results of the 2^3 factorial design. The design indicated that there were no main effects or interactions on the French uranine method from relative humidities up to 80%, temperature up to 90°C, and NO_x concentrations up to 10,000 ppm. Based on this satisfactory performance, the uranine method was selected to determine the particle retention efficiency of the Atmosphere Protection System HEPA filter banks.

The results under the in-plant demonstration are limited due to operation of the waste calciner. We recommend further measurements at the APS filters to study the effect of NO_x and uranine particle size on the French uranine method.

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